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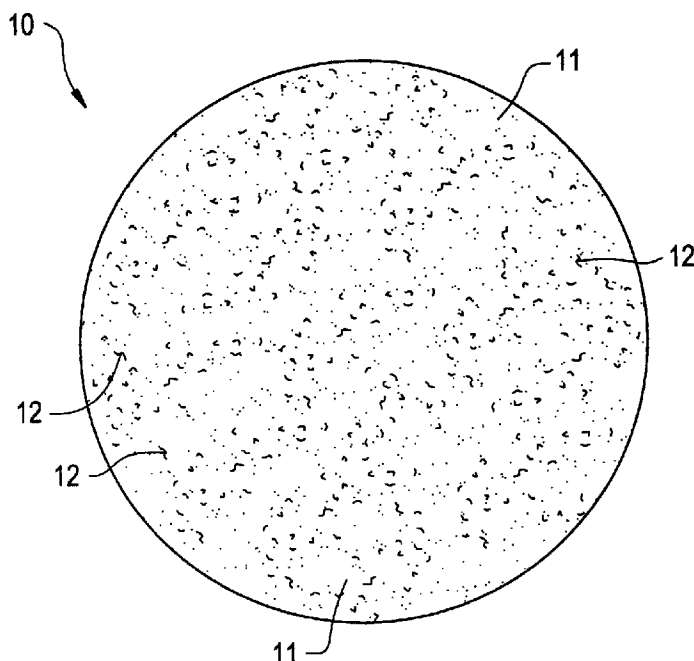
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(54) Title: POLYMER COMPOSITE STRUCTURES USEFUL FOR CONTROLLED RELEASE SYSTEMS



(57) Abstract: Solid polymer composites comprising a matrix of at least one water-soluble polymer and at least one chemically distinct species forming a dispersed phase are disclosed. Such composites are suitable for controlled release systems in, for example, laundry and dishwashing applications. Methods of making the solid polymer composites are also disclosed.



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## POLYMER COMPOSITE STRUCTURES USEFUL FOR CONTROLLED RELEASE SYSTEMS

The present invention relates to a polymer composite useful for controlled release of an active ingredient.

5        Traditional methods of delivery of detergents and/or additive compositions for use in dishwashing applications or laundry applications include solid granulated particles, pouches or sheets. In the case of laundry applications, for example, the detergent, be it in granulated, liquid, pouch or sheet form, is added to the washing machine, together with the clothing to be washed.

10        When the detergent is in granulated form, often there are problems with dissolution of the detergent. In other words, the solid granules remain in solid form throughout the wash cycle, thereby minimizing the effectiveness of the detergent, and often leaving a residue on the clothing.

15        When the detergent is in liquid form, all of the detergent and/or additives are released without control into the washing machine or dish washer, which is not necessarily desirable. Also, liquids do not permit segregation of the various ingredients.

20        U.S. Patent Nos. 5,783,541, 5,160,654, 5,116,524, 5,110,640, 4,925,586, 4,806,261, and 4,416,791 all teach various detergent packages in the form of sachets or pouches. In all of these patents, a granular detergent is contained within a pouch formed by a sheet or sheets of either water-soluble or water insoluble material. However, in all of these cases, the release of the detergent into the washing machine or dishwasher is delayed until either the water-soluble sheet dissolves, or until a seal breaks to release the detergent. Once the detergent is released, dissolution of the detergent actives occurs without control.

25        Attempts at varying the release times of detergents and additives is described in U.S. Patent Nos. 4,776,455, 4,410,441, which teach multicompartment sachets, wherein the contents of the separate compartments are released at different times. However, in both of these patents, once the compartment is opened by dissolution of the walls surrounding the compartment, the contents within are released without control.

30        Likewise, U.S. Patent Nos. 5,202,045, 5,053,157, 5,030,375, 4,919,835, 4,865,755, 4,652,390, 4,532,063, 4,095,946, and 4,170,565 teach the use of sheet-like materials coated with detergent materials, again once when this article is place into the wash bath the detergent actives are released with out control which may be undesirable.

EP 0 164 703 B1 claims a washing additive comprising a mixture of detergent ingredients and, optionally, auxiliaries encapsulated in organic, flexible, film-forming water-soluble polymers. The detergent ingredients are not on the surface of the polymer, but rather are encapsulated within the polymer, such that the detergent ingredients are not released upon  
5 introduction to the washing machine. Such an additive thus presents the same problem as the pouch or sachet, in that one must wait for dissolution of the polymer before releasing the ingredients into the wash.

It would be an advance in the art to provide a mechanism whereby release of an active ingredient can be controlled in a variety of ways, as opposed to a simple delayed release.

10 In one aspect, the present invention is a solid polymer composite comprising a matrix of at least one water-soluble polymer and at least one chemically distinct species forming a dispersed phase.

In a second aspect, the present invention is a method of making a solid polymer composite comprising mixing at least one water soluble polymer with at least one chemically  
15 distinct species as the dispersed phase to form a mixture and shaping the mixture into a desired polymer composite structure.

In a third aspect, the present invention is a controlled release system comprising at least one polymer composite and optionally other non-composite polymers, active ingredients or combinations thereof.

20 The controlled release systems of the present invention are useful for numerous applications, including delivery of detergents and/or additive compositions for use in dishwashing, dilutable hard surface cleaner or laundry applications, and addition of, for example, initiators or chain transfer agents to chemical processes, as well as addition of swimming pool chemicals, fish tank chemicals, water treatment chemicals and delivery of  
25 drug or other actives from pills. It will be obvious to one skilled in the art that a multitude of controlled release systems can be generated providing a number of release profiles. A preferred embodiment is to have the active ingredient intimately incorporated into the polymer matrix, the active ingredient can be released as soon as the polymer composite is introduced to the laundry, dishwashing, or other system, thereby avoiding the delay associated with the  
30 pouch/sachet structures of the prior art. As the polymer dissolves, the active ingredient is continuously released, which allows much better control and is unlike the pouch/sachet/sheet

systems of the prior art. Moreover, the methods for making such polymer composites are simplified over the methods used to make the pouch/sachet/sheet systems of the prior art.

The following Figures are examples of, but not limited to, controlled release systems contemplated by this invention.

5        Figure 1 depicts a front view of a pellet of the present invention, with an active ingredient continuously dispersed throughout a polymer matrix.

Figure 2 depicts a front view of a pellet with an active ingredient dispersed in an isolated region of a polymer matrix.

10       Figure 3 depicts a front view of a polymer composite multilayer film of the present invention.

Figures 4A-4B depict cross-sectional views of a polymer composite pouch of the present invention.

Figures 5-9 are charts illustrating the benefits of the present invention.

15       The polymer used for the polymer matrix of the present invention is preferably a water-soluble polymer. Suitable water soluble polymers include polyvinyl alcohol and copolymers thereof, partially hydrolyzed polyvinyl acetate, alginates, cellulose ethers such as carboxymethylcellulose and methylcellulose, polyacrylates, polyethylene oxide, polyethylene glycol, starch, sulfonated polyesters, sulfonated polystyrene and copolymers thereof, polyacrylamides, polyvinylpyrrolidinone, polymaleic anhydride, and some modified cellulose materials such as cellulose acetate.

20       As used herein, the term "chemically distinct species" means any ingredient that can be dispersed within the polymer such that it interrupts the structure of the polymer in order to create more surface area. Such interruption enables the polymer to dissolve faster than the polymer would dissolve without the chemically distinct species. The chemically distinct species used for the present invention can be one or more active ingredients, one or more inert actives, and combinations thereof.

25       Active ingredients useful for the present invention include surfactants, soaps, builder salts, chelants, zeolites, peroxide bleaching agents, enzymes, enzyme stabilizers, enzyme co-factors, dissolution aids, hydrotropes, foam suppressors, anti-redeposition aids, polymeric builders, soil dispersants, crystal growth inhibitors, fabric whiteners, fabric brighteners, disinfecting agents, dye transfer inhibitors, fabric softeners, fragrance, initiators, solvents, chain transfer agents, fertilizers, insecticides, fungicides, pesticides, nutrients, disinfectants,

preservatives, emollients, buffering agents, antibacterial agents, vitamins, alcohols, salicylic acid, sodium hypochlorite, quaternary amine salts, and combinations and encapsulants thereof.

Inert actives useful for the present invention include calcium carbonate, sodium sulfate, sodium chloride, and clays. In a preferred embodiment, the chemically distinct species is at least one active ingredient.

Figure 1 depicts one embodiment of the present invention. As shown, a pellet comprises a water-soluble polymer matrix 11 and at least one active ingredient 12 dispersed continuously throughout the matrix 11.

As illustrated in Figure 2, the polymer composite pellet of the present invention can be combined with a noncomposite polymer to form a multilayer "core/sheath" pellet 20. The inner layer or "core" 21 comprises a polymer matrix 22 and at least one chemically distinct species 23. Outer layer or "sheath" 24 comprises a polymer. Polymer matrix 22 and polymer 24 may be the same material, or they may be different materials.

In another embodiment, the polymer composite of the present invention can be elongated and thereby have the shape of fibers.

In yet another embodiment, the polymer composite of the present invention can be elongated and widened, such that it has the shape of a film. Advantageously, the film can be a monolayer film, wherein one or more chemically distinct species are dispersed throughout the film, or the film can be a multilayer film.

As illustrated in Figure 3, the polymer composite films of the present invention can be combined to form a multilayer film 30. Multilayer film 30 is formed from at least a first layer 31 and a second layer 34. First layer 31 comprises a polymer matrix 32 and at least one chemically distinct species 33. Second layer 34 comprises a polymer matrix 35 and at least one chemically distinct species 36. Polymer matrices 32 and 35 may be the same material, or they may be different materials. Likewise, species 33 and 36 may be the same material or combination of materials, or they may be different materials.

If desired, the films can also be manipulated so as to form a pouch. Figure 4A illustrates a pouch 40 made from a polymer composite film of the present invention. As shown in Figure 4A, the pouch 40 can be filled with an additional active ingredient 41. In this embodiment, once pouch 40 dissolves, thereby slowly and continuously releasing the active ingredient therein, active ingredient 41 will be released immediately.

Figure 4B also illustrates pouch 40, which is filled with pellets 10 of the present invention. In this manner, once pouch 40 dissolves, pellets 10 will be released, and the active ingredient contained in pellets 10 will thereafter be continuously and slowly released. If desired, pellets 20, containing a different active ingredient than pellets 10, can also be  
5 contained within pouch 40, thereby releasing the active ingredients contained in pellets 20.

The amount of the chemically distinct species contained in the polymer matrix will depend upon the identity of the chemically distinct species and will also depend upon the length of time over which release of the active ingredient is desired as well as the ultimate size of the matrix. For example, the loading level of detergent surfactant for a laundry application  
10 is likely to be greater than the loading level for a detergent whitener. Moreover, the release of detergent actives for a laundry application is likely to be over a relatively short time period as compared to the release of a swimming pool chemical. Preferably, however, the loading of the chemically distinct species is from 0.1 weight percent to 80 weight percent, based on the total weight of the composite.

The release rate of the chemically distinct species in the dispersed phase can be controlled by selection of polymer type, crosslinking of the polymer matrix, molecular weight of the polymer, level of dispersed phase incorporated into the composite, geometry of the composite, or combinations thereof. In a preferred embodiment, the composite shows utility  
15 in, but is not limited to, controlled release of detergent additives. In this application the release rate of the dispersed phase is from 100g per minute to 0.1g per 10 minutes.

To make the polymer composites of the present invention, the water soluble polymer and at least one chemically distinct species are mixed together to form a mixture. The mixture is then shaped into a desired polymer composite structure.

The mixing of the composite can be done in the dry state with sufficient agitation to  
25 obtain thorough distribution of ingredients. Additionally, the mixing of the composite can be done by subjecting the ingredients to shearing stresses at elevated temperature with or without the addition of liquid ingredients with or without vacuum. The shaping of the composite can be performed using any desired technique, such as, for example, compression molding, extrusion, solution casting, pelletizing, wet spinning, dry spinning and meltspinning, melt  
30 blowing. The resulting composite can be in the shape of a fiber, multicomponent fiber, film, multilayer film, filled pouch, non-woven, pellets, multicomponent pellets, tablet or combinations thereof.

The size of the resulting tablets, pellets, fibers or films is not critical, as long as the pellets, fibers or films display the desired controlled release properties. In addition to the polymer composite, the controlled release systems of the present invention can include at least one other non-composite polymer or one or more additional active ingredients.

5           In an embodiment of the present invention, a laundry composition can be designed having surfactants with different HLBs (Hydrophilic Lipophilic Balance), such that the HLB changes as a function of time in the wash cycle. Such an embodiment is advantageous in a laundry application, because surfactants with different HLBs work on different types of laundry soils. In such an embodiment, a surfactant of low HLB can be released relatively  
10           quickly, whereas a surfactant of high HLB can be delayed and released slowly as the wash cycle progresses. As the high HLB surfactant is released the overall HLB of the surfactant system in the laundry bath would gradually change from low to high, allowing for effective cleaning on different types of soils. Other embodiments are also contemplated for laundry applications. For example, a pouch made from a composite of the present invention can  
15           contain pellets made using the composite of the present invention. The active ingredient in the pouch may be a detergent, whereas the active ingredient in the pellets can be a bleaching agent, a fabric softener, combinations thereof, or any other desired active. In this manner, the detergent will be slowly released into the wash cycle, and thereafter, the fabric softener and/or other active will be released.

20           In another embodiment of the present invention, the polymer composite, in the form of a non-woven sheet, can be used as the top sheet in a disposable diaper. When the diaper is insulted, skin care actives can be released. Such actives include but are not limited to buffering agents, such as sodium bicarbonate and sodium citrate; emollients, such as glycerin, sorbitol, glycerol, ethylene glycol, and propylene glycol; and antibacterial agents, such as  
25           quaternary amine salts, benzalkonium chloride, chlorhexidine, those marketed under the tradename DOWICIL by The Dow Chemical Company, and triclosan.

          Other embodiments of the present invention, having the form of a non-woven sheet, include those that require the non-woven to be wetted before use to release the active ingredient or ingredients. Examples of such embodiments are baby wipes, wherein the actives  
30           include surfactants, emollients; fragrances, buffering agents and/or antibacterial agents; facial cleansing tissues and make-up removal tissues, wherein the actives include emollients, surfactants, vitamins, alpha hydroxyl acids and/or salicylic acid; and hard surface cleaners and

disinfectants, wherein the actives include surfactants, chelants, solvents, sodium hypochlorite and/or quaternary amine salts.

In a further embodiment of the present invention, fibers of multiple composition can be prepared to separate cleaning components which are not compatible with one another. The active fibers can be added to a nonwoven or composite structure that serves as a cleaning device or a scrubbing device for multiple uses around the home. The life of the scrubbing device can be tailored to be long or short (multiple uses versus one time use) by the selection of the composition of the active and inactive fibers making up the cleaning device.

In still another embodiment of the present invention, a polymer composite structure in the form of a film can be prepared which serves as the active dispensing agent in a cleaning structure. The cleaning structure can be in the form of a sponge, flexible or rigid abrasive pad, brush, or other general or special purpose cleaning device. The film may be coated onto the cleaning structure, laminated to it or impregnated into it. In each case, the film delivers the cleaners to the surface to be cleaned.

Yet another embodiment of the present invention is a convenience pack useful for the manufacture of formulated products. Accurate addition of small quantities of solids to batch formulation can be difficult. In this embodiment, a pre-weighed quantity of active is contained in a solid polymer composite article, such as a filled pouch, which is added to the formulation. The article dissolves and the active is delivered to the formulation.

A similar embodiment is the use of the polymer composite of the present invention in the dilution of active agricultural ingredients at the point of use. In this embodiment, a pre-weighed quantity of active such as fertilizer, pesticide, or fungicide is contained in an article that is dissolved in water to make the desired concentration of actives. This eliminates the need to weigh actives, yet still provides an accurate concentration.

Yet a further embodiment of the present invention is the controlled release of agricultural actives, such as fertilizers, pesticides, fungicides, and/or nutrients, from solids that are located in the proximity of plants. The actives are released upon contact with water. Depending on the nature of the controlled release system, the release rate of the active may be short, on the order of minutes, or release may be longer, on the order of days. The release rate of each active can be controlled separately. In this embodiment, the controlled release system can be in the shape of pellets that are sprinkled over the plants, or the controlled release system can be in the shape of a film that is laid below the ground surface. Advantageously, a

composite film could contain a non-water soluble layer that prevents weed growth, while the water-soluble polymer composite layer or layers deliver the actives.

Controlling the release of volatile fragrances and perfumes can also be achieved through the use of the present invention. To achieve the controlled release of fragrances or perfumes, such fragrances and perfumes can be added directly to the polymer, or the fragrances and perfumes can be mixed with surfactants and incorporated into the water soluble polymer, or the fragrances and perfumes can be adsorbed onto inert particles such as clays and zeolites and then dispersed within the water soluble polymer matrix. This provides immediate and long lasting perfume effect. Additionally, this provides for release of perfumes upon dissolution in water, the rate of which can be controlled by the properties of the water soluble polymer matrix.

Yet another embodiment is the use of the present invention for the delivery of automatic dish washing detergent actives. In such an embodiment, as with laundry applications, incompatible agents can be incorporated into the detergent composition. Thus, for example, an enzyme and bleach can be incorporated into the matrix and released at different times in the wash cycle.

Yet another embodiment is the use of the present invention as a dilutable hard surface cleaner. In such an embodiment, the controlled release article, possibly in the shape of a pellet, is diluted in a bucket to the appropriate concentration of actives. Actives could include those listed above for the laundry application.

Other embodiments of the present invention are also contemplated. The following examples are for illustrative purposes only and are not intended to limit the scope of the claimed invention. Percentages are in weight percents unless otherwise stated.

## Examples

The experimental results are shown in Figures 5-9. For all of the examples that follow, the release profile of the active ingredient or polymer was measured using electrical conductivity (YSI Scientific Model 35, Conductance Meter) as a function of time as the active ingredient, polymer, composite or controlled release system was dissolved into one liter of deionized water at 21°C. The dissolution was carried out in a 6-place Terg-o-tometer (U.S. Testing, Hoboken, NJ) at 100rpm. The conductivity measurements were reported as normalized readings to assist in comparative analysis.

Comparative Example A

The rate of release of 1 gram a detergent polymer (Acusol 479ND, Rohm and Haas, Philadelphia, PA) powder was measured. The results are plotted in Figure 5.

5 Examples 1-3

Polymer composite films weighing 3.3 grams were formed, wherein the polymer matrix was polyethylene oxide (POLYOX N-10, Union Carbide, Danbury, CT) and the dispersed phase was a detergent polymer powder (30 percent by weight). The PEO and detergent polymer powder were mixed using a ball mill and then compression molded

10 (Pasadena Hydraulics Inc.) according to the following conditions:

Example	Temperature (°C)	Warm up time (min)	Pressure (ram tons)	Time at pressure (min)	Film thickness (mils)
1	100	3	0	0	25
2	100	10	0	0	16.9
3	100	10	5	2	9.5

Figure 5 shows the results, as compared to those of Comparative Example A. As shown, the polymer composite films allows for a controlled release as compared to the

15 detergent polymer powder alone.

Comparative Example B

The rate of release of 1 gram of a fully formulated powdered laundry detergent (Tide Powder, Procter and Gamble, Cincinnati, OH) was measured. The results are plotted in Figure

20 6.

Examples 4-5

Polymer composite fibers were formed, wherein the polymer matrix was polyethylene oxide (PEO) and the dispersed phase was Tide powder laundry detergent. A mixture

25 containing 70 percent by weight percent PEO and 30 percent by weight of Tide was prepared using a Haake Rheodrive Model 7500 (Paramus, NJ) Mixing conditions were 40 rpm at 110°C for 15 minutes after a 5 minute warm-up period. The mixture was then extruded into fibers using a capillary rheometer (Rheograph 2003, Gottfert Inc., Rockhill, SC) with a barrel

temperature of 100°C. The fiber extruded through a 0.5 mm die had a thickness of 18.5 mils, whereas the fiber extruded through a 1.0 mm die had a thickness of 48 mils.

The release rates of the active from 3.3 grams of fibers were then measured. The results are plotted in Figure 6. As shown in Figure 6, the rate of release of the Tide can be controlled easily by fiber geometry.

#### Comparative Example C

The rate of release of 1 gram a detergent polymer (Acusol 479ND, Rohm and Haas, Philadelphia, PA) powder was measured. The results are plotted in Figure 7.

#### Examples 6-8

Mixtures of Acusol/PEO (containing 30 percent by weight of Acusol) were formed into 3.3 gram films according to the method described for Examples 1-3. The resulting film thicknesses for Examples 6, 7 and 8 were 12.5 mils, 16.7 mils and 16.9 mils, respectively.

The release rates were measured according to the procedures described for Examples 1-3. The results are shown in Figure 7.

#### Comparative Example D

A film formed from PEO alone was formed according to the method described for Examples 1-3. The resulting film thickness was 12.5 mils. The dissolution rate of a 3.3 gram film was measured according to the procedures described for Examples 1-3. The results are shown in Figure 7. As shown, the film formed from PEO only dissolved more slowly than films containing a chemically distinct species.

#### Comparative Example E

The rate of release of 1 gram of an anionic deterative surfactant, Calsoft F90(Pilot Chemical Co., Santa Fe Springs, CA) powder was measured. The results are plotted in Figure 8.

#### Example 12

A Calsoft F90/PEO film (containing 30 percent by weight of Calsoft 90) was formed according to the method described for Examples 1-3. The resulting film thickness was 11.2

mils. The release rate of a 3.3 gram film was measured according to the procedures described for Examples 1-3. The results are shown in Figure 8.

#### Comparative Example F

5 A film formed from PEO alone was formed according to the method described for Examples 1-3. The resulting film thickness was 10 mils. The dissolution rate of the 3.3 gram film was measured according to the procedures described for Examples 1-3. The results are shown in Figure 8. As shown, the film formed from PEO only dissolved more slowly than a film containing a chemically distinct species.

#### Example 13

10 A polymer composite pouch was formed by first forming two films made from PEO only according to the method described for Examples 1-3 wherein the compression molding conditions were 100°C, 5 minutes warm up time, no pressure. The resulting film thicknesses were 9.5 mils for each film. The weight of the two films was 2.3 grams. 1 gram of Tide powder was placed between the two films, and the edges were then heat sealed with a hand held rotary heat sealer. The release rate was measured according to the procedures described for Examples 1-3. The results are shown in Figure 9. This shows the delay that unavoidable occurs in pouches of the current art.

#### Example 14

20 A Tide/PEO film was formed according to the method described for Examples 1-3. The resulting film thickness was 11.4 mils. The release rate of the 3.3 gram film was measured according to the procedures described for Examples 1-3. The results are shown in Figure 9. This shows an advantage to the existing art as delivery can, if desired, start almost immediately.

#### Example 15

30 Polymer composite fibers were formed, wherein the polymer matrix was polyethylene oxide (PEO) and the dispersed phase was a powdered or liquid general purpose cleaner or cleaning solvent. The mixture was prepared in a ratio of cleaner to PEO that resulted in a formulation that was extruded into fibers. Fibers of multiple composition were prepared to

separate cleaning components which were not compatible with one another. The active fibers were added to a nonwoven or composite structure that served as a cleaning device or a scrubbing device for multiple uses around the home. The life of the scrubbing device can be tailored to be long or short (multiple uses versus one time use) by the selection of the  
5 composition of the active and inactive fibers making up the cleaning device.

WHAT IS CLAIMED IS:

1. A solid polymer composite comprising a water-soluble polymer matrix and at least one chemically distinct species forming a dispersed phase.

2. The polymer composite of Claim 1 wherein the polymer matrix comprises at least one water-soluble polymer.

3. The polymer composite of Claim 1 wherein the dispersed phase is released as the water-soluble polymer matrix is dissolved.

4. The polymer composite of Claim 1 wherein the release rate of the dispersed phase can be controlled by selection of polymer type, crosslinking of the polymer matrix, molecular weight of the polymer, level of dispersed phase incorporated into the composite, geometry of the composite, or combinations thereof.

5. The polymer composite of Claim 1 wherein the dispersed phase is selected from the group consisting of active ingredients, inert additives, and combinations thereof.

6. The polymer composite of Claim 1 wherein the active ingredient is selected from the group consisting of surfactants, soaps, builder salts, chelants, zeolites, enzymes, enzyme stabilizers, enzyme co-factors, dissolution aids, hydrotropes, foam suppressors, anti-redeposition aids, polymeric builders, soil dispersants, crystal growth inhibitors, fabric whiteners, fabric brighteners, dye transfer inhibitors, fabric softeners, fragrance, initiators, chain transfer agents, fertilizers, insecticides, fungicides, pesticides, disinfectants, preservatives, emollients, buffering agents, antibacterial agents, vitamins, alphahydroxy acids, salicylic acid, sodium hypochlorite, quaternary amine salts, solvents and combinations and encapsulants thereof.

10. A method of making a solid polymer composite comprising: mixing at least one water soluble polymer with at least one chemically distinct species as the dispersed phase to form a mixture; and shaping the mixture into a desired polymer composite structure.

11. The method of Claim 10 wherein the release rate of the dispersed phase can be controlled by selection of polymer type, crosslinking of the polymer matrix, molecular weight of the polymer, level of dispersed phase incorporated into the composite, geometry of the composite, or combinations thereof.

12. The method of Claim 10, wherein the chemically distinct dispersed phase is selected from the group consisting of active ingredients, inert additives, and combinations thereof.

13. The method of Claim 10 wherein the active ingredient is selected from the group consisting of surfactants, soaps, builder salts, chelants, zeolites, enzymes, enzyme stabilizers, enzyme co-factors, dissolution aids, hydrotropes, foam suppressors, anti-redeposition aids, polymeric builders, soil dispersants, crystal growth inhibitors, fabric  
5 whiteners, fabric brighteners, dye transfer inhibitors, fabric softeners, fragrance, initiators, chain transfer agents, fertilizers, insecticides, fungicides, pesticides, disinfectants, preservatives, emollients, buffering agents, antibacterial agents, vitamins, alphahydroxy acids, salicylic acid, sodium hypochlorite, quaternary amine salts, solvents and combinations and encapsulants thereof.

10 14. The method of Claim 10 wherein the mixing of the composite is done in the dry state with sufficient agitation to obtain thorough distribution of ingredients.

15 15. The method of Claim 10 wherein the mixing of the composite is done by subjecting the ingredients to shearing stresses at elevated temperature with or without the addition of liquid ingredients with or without vacuum.

16. The method of Claim 10 wherein the shaping of the composite is done via compression molding, extrusion, solution casting, wet spinning, dry spinning, meltspinning, melt blowing.

20. A controlled release system comprising at least one composite of Claim 1 and optionally other non-composite polymers, active ingredients or combinations thereof.

20 21. The controlled release system of Claim 20 wherein the release rate of the dispersed phase from each composite can be separately controlled by selection of polymer matrix type, crosslinking of the polymer matrix, molecular weight of the polymer matrix, level of dispersed phase incorporated into the composite, geometry of the composite, non-composite polymer type, non-composite polymer molecular weight, non-composite polymer  
25 geometry, design of the controlled release system or combinations thereof.

22. The controlled release system of Claim 20 comprising at least one composite of Claim 1, the system having the shape of a fiber, multicomponent fiber, film, multilayer film, filled pouch, non-woven, tablets, multicomponent tablets, pellets, multicomponent pellets or combinations thereof.

30 23. The controlled release system of Claim 20 comprising at least one composite of Claim 1 and at least one other non-composite polymer the system having the shape of a

multicomponent fiber, multilayer film, filled pouch, non-woven, multicomponent pellets, multicomponent tablets, or combinations thereof.

24. The controlled release system of Claim 20 comprising at least one composite of Claim 1 and at least one active ingredient the system having the shape of a multicomponent  
5 fiber, multilayer film, filled pouch, non-woven, multicomponent tablets, multicomponent pellets or combinations thereof.

25. The controlled release system of Claim 20 comprising at least one composite of Claim 1, at least one other non-composite polymer and at least one active ingredient the system having the shape of a multicomponent fiber, multilayer film, filled pouch, non-woven,  
10 multicomponent pellets or combinations thereof.

26. The controlled release system of Claim 20 wherein the active ingredient is selected from the group consisting of surfactants, soaps, builder salts, chelants, zeolites, enzymes, enzyme stabilizers, enzyme co-factors, dissolution aids, hydrotropes, foam suppressors, anti-redeposition aids, polymeric builders, soil dispersants, crystal growth  
15 inhibitors, fabric whiteners, fabric brighteners, dye transfer inhibitors, fabric softeners, fragrance, initiators, chain transfer agents, fertilizers, insecticides, fungicides, pesticides, disinfectants, preservatives, emollients, buffering agents, antibacterial agents, vitamins, alphahydroxy acids, salicylic acid, sodium hypochlorite, quaternary amine salts, solvents and combinations and encapsulants thereof.

27. The use of the controlled release system of Claim 20 to clean laundry in a washing machine.

28. The use of the controlled release system of Claim 20 to clean dishes in a dishwasher.

29. The use of the composite structure of Claim 25 as a top sheet in a diaper, as a  
25 baby wipe, as a facial cleansing tissue or as a hard surface cleaner.

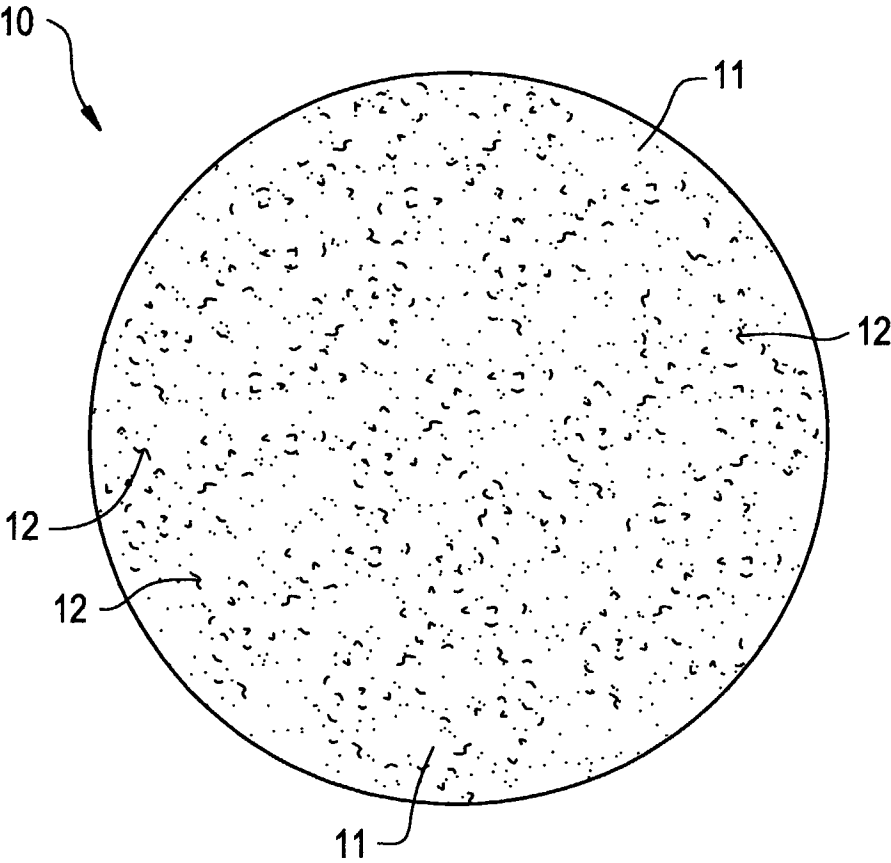
30. The use of the controlled release system of Claim 20 as a cleaning structure, wherein the cleaning structure has the form of a sponge, flexible or rigid abrasive pad, brush or scrubbing device.

31. The use of the controlled release system of Claim 20 in agricultural  
30 applications.

32. The use of the controlled release system of Claim 20 in the manufacture of formulated products.

33. The use of the controlled release system of Claim 20 in the release of volatile fragrances or perfumes.

FIG. 1



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FIG. 2

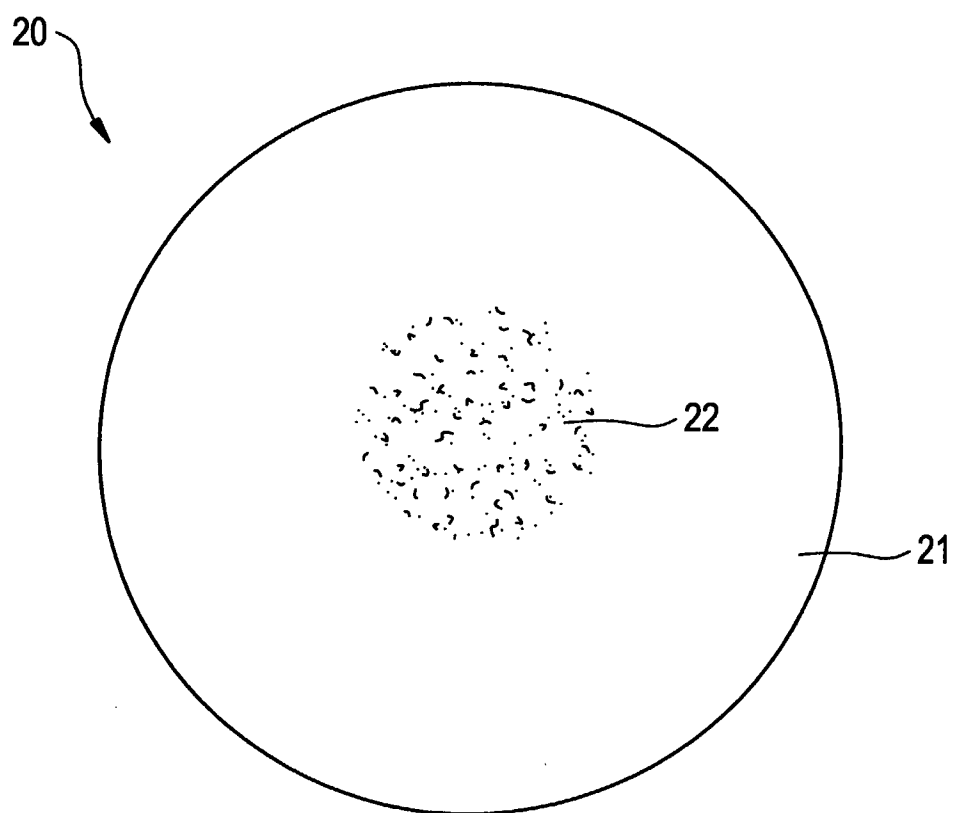


FIG. 3

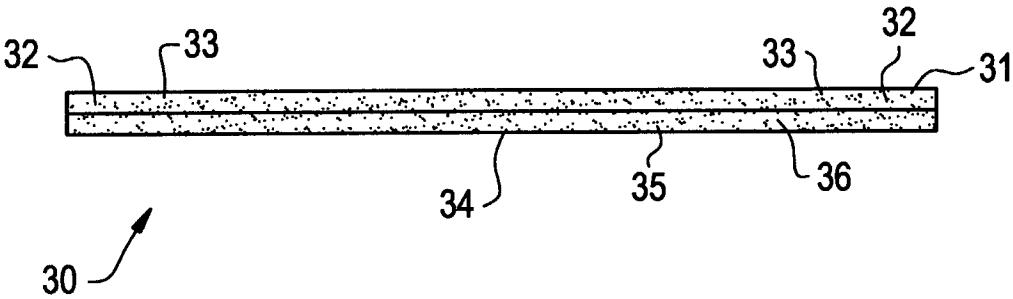


FIG. 4A

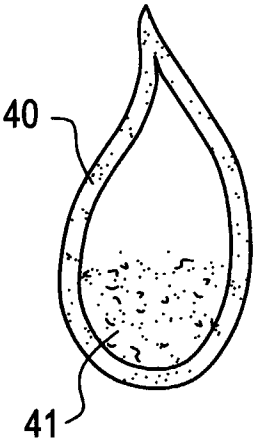
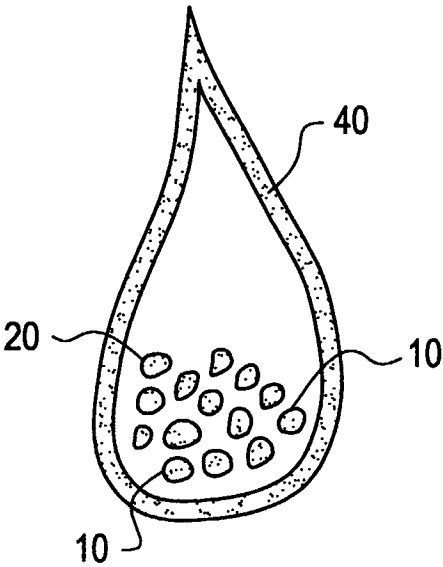


FIG. 4B



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FIG. 5

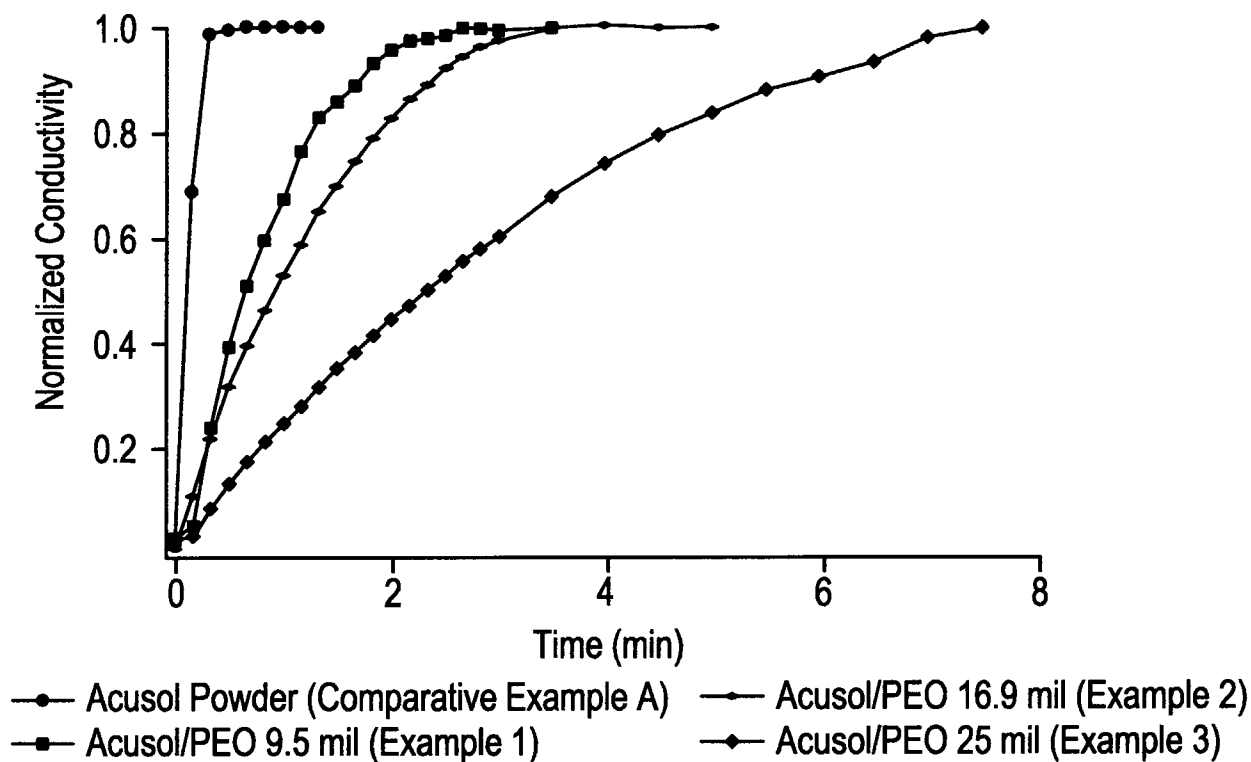
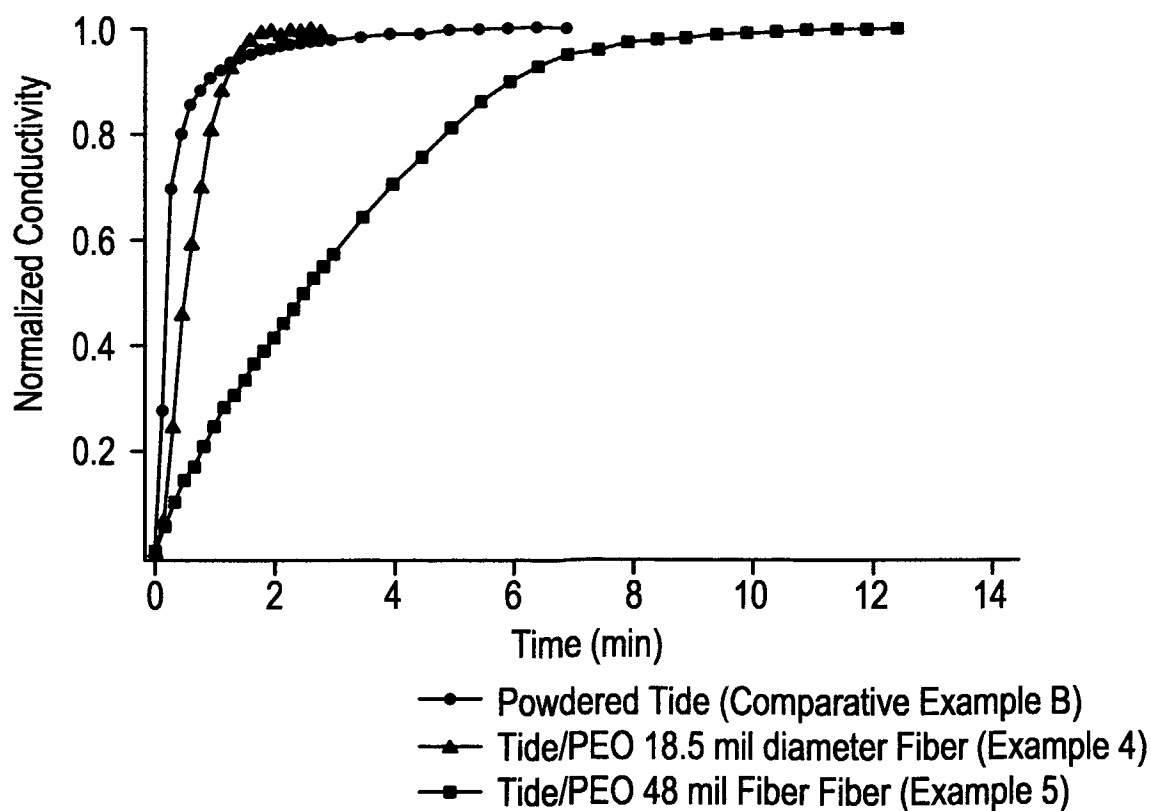


FIG. 6



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FIG. 7

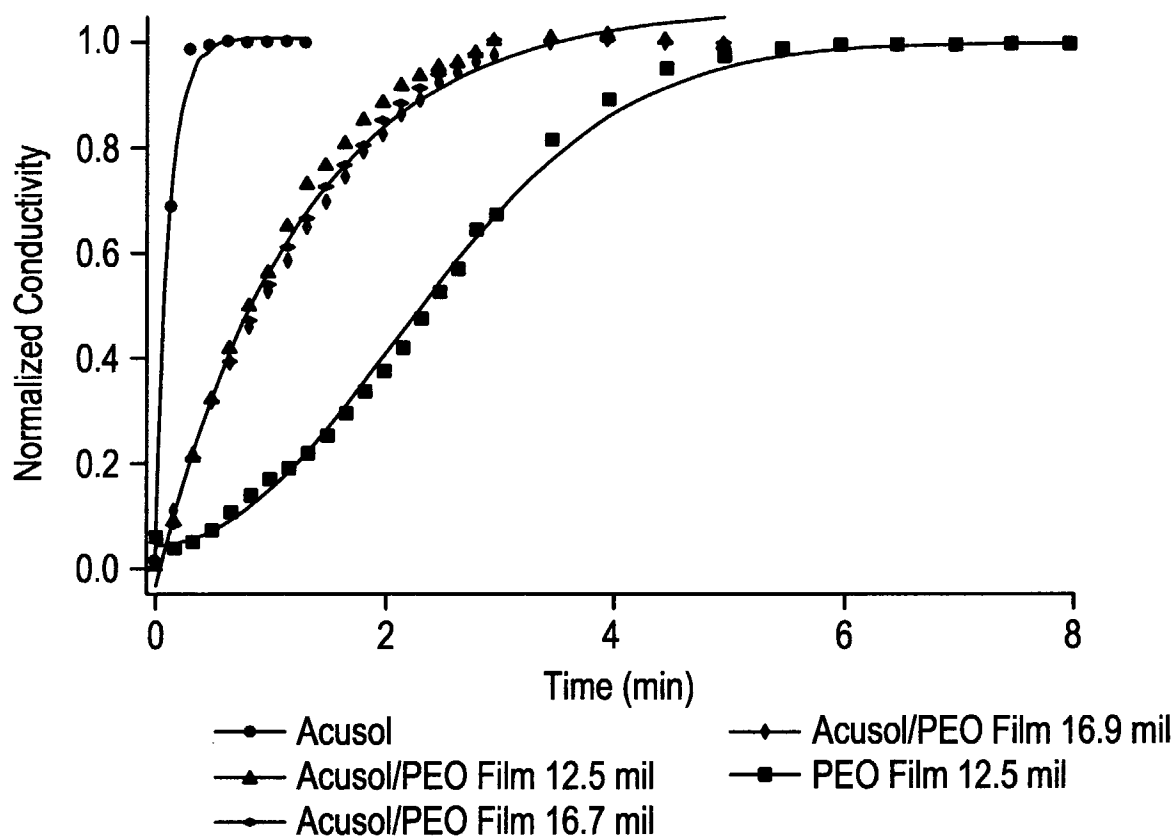


FIG. 8

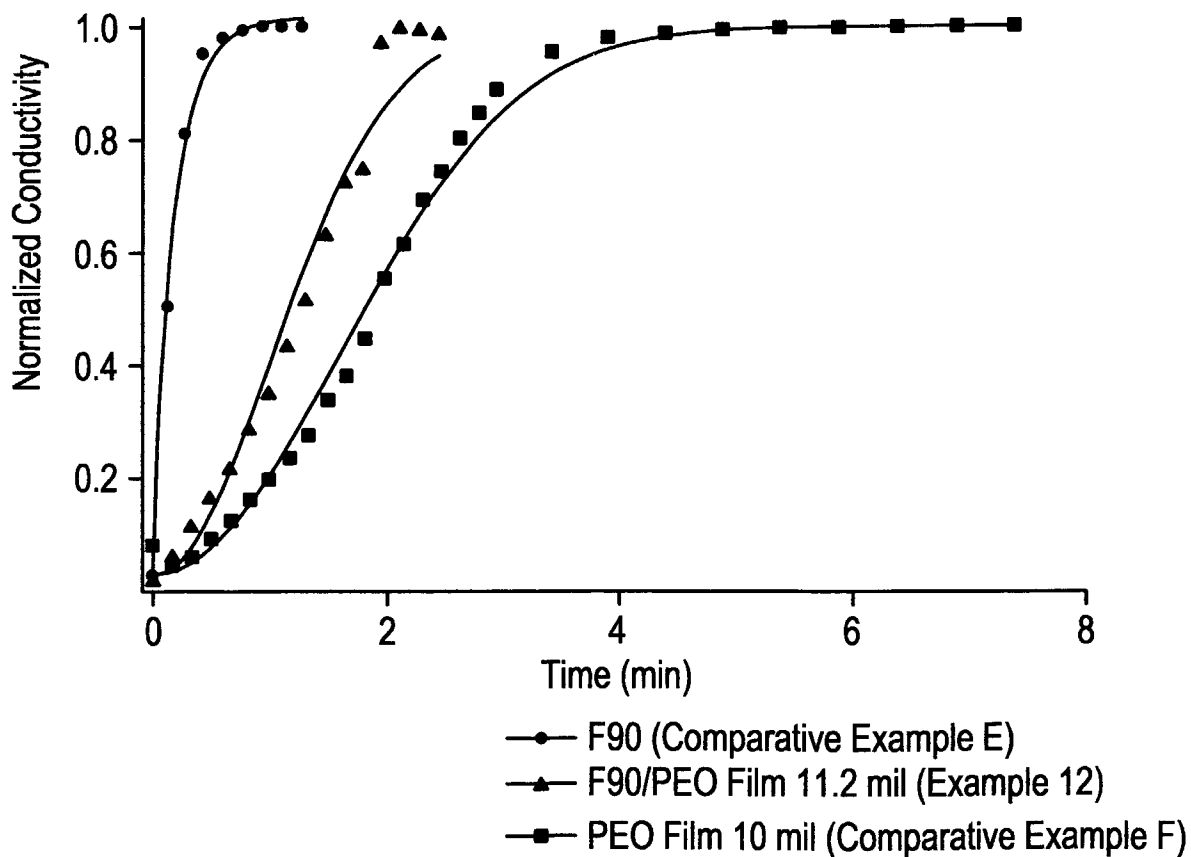
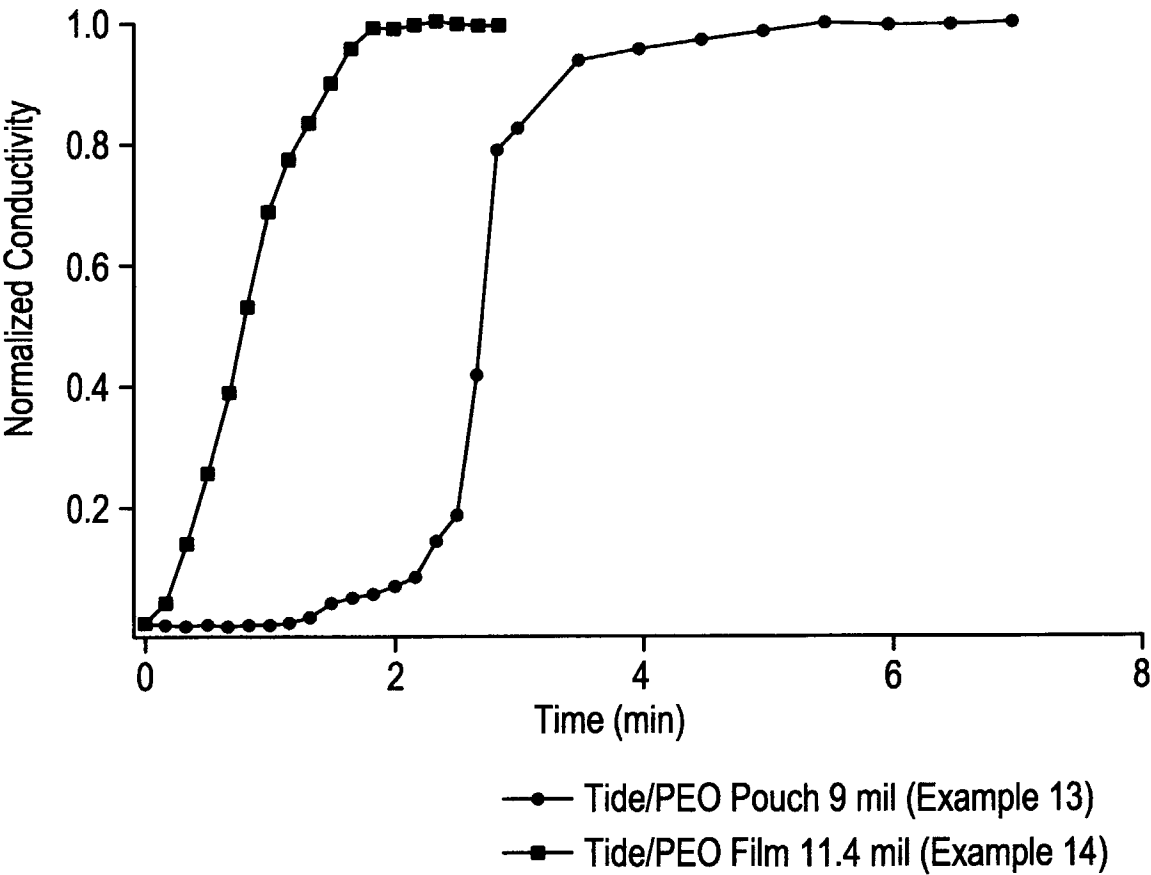


FIG. 9



## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C11D17/04 C11D3/37 A01N25/10 A01N25/34 C11D3/50

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C11D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

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☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

24 January 2002

Date of mailing of the international search report

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## INTERNATIONAL SEARCH REPORT

Inter - 1al Application No

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